

RESERVE COPY PATENT SPECIFICATION

615,425



Application Date: July 30, 1945.

No. 19547/45.

Complete Specification Left: Feb. 5, 1946.

Complete Specification Accepted: Jan. 6, 1949.

Index at acceptance:—Classes 1(i), F4(a: b: c), L(1: 3); 2(iii), C3a13b2, N; 23, K2; 81(i), I; 82(i), Q; and 91, C1(a3: c), C2(a1: b: c: e).

PROVISIONAL SPECIFICATION

Improvements in Method of Contacting Fluids One with Another

I, JAMES GEORGE FIRE, M.Sc., Ph.D., F.R.I.C., Consulting Chemist and Chartered Patent Agent, of Bank Chambers, 329, High Holborn, London, W.C.1, a

British Subject, do hereby declare the nature of this invention (as communicated to me from abroad) to be as follows:—

The present invention relates to methods of contacting fluids and fluidized materials and to apparatus in which to execute the same. More particularly, the present invention pertains to the treatment of mixtures of fluids in which advantage is taken of the formation of discrete phases.

It is an object of this invention to obtain intimate contact between fluids or fluidized materials of dissimilar physical and/or chemical properties. It is a further object to obtain efficient contact in relatively short periods of time. It is still another object to obtain high contact efficiency in relatively short distances of contact. It is a still further object to form a discrete discontinuous phase of uniform pattern in a continuous phase. Another object is to form a discontinuous phase whose discrete fluid particles are of a predetermined substantially uniform size. Still another object is the obtainment of a dispersed phase of materials of finely divided fluid or fluidized character. A further object is the development of a method of contact which will function efficiently independently of varying physical or chemical properties of the materials being treated and of equipment in which said method can be carried out. Another object is the creation of an apparatus capable of achieving various purposes with little or no change in fundamental design. Still other objects include the development of improved separation technique, the formation of novel rules of extraction; the use of simple but highly efficient equipment, and the processing of large quantities of materials in relatively small

mechanical units possessed of high throughput capacities.

These and other objects are within the purview of the present invention and will be elaborated upon in more specific portions of the specification.

The fluid or fluidized material comprehended by this invention, hereinafter called "fluids," embraces liquids, gases, vapors, powders and finely divided bodies, which are capable of forming a multiphase system or, more specifically, a dispersion of at least one type of material in a continuous phase of the other, which phases remain discrete throughout the major portion of the contact zone. Illustrative of the general types of heterogeneous systems contemplated are: gas-solid, gas-liquid, liquid-gas, liquid-liquid, liquid-solid, liquid-liquid-solid, liquid-gas-solid, gas-liquid-solid, liquid-liquid-gas, liquid-liquid-liquid and gas-liquid-liquid systems wherein the first-named constituent comprises a continuous phase and the second constituent (with the third) comprises a dispersed phase or phases. Each phase may comprise a plurality of components such as a mixture of solvents of differing capacities or different degrees of selectivity, mixtures containing diluents for reduction of viscosity, for change in surface tension or interfacial tension, for alteration of internal density, etc. The fluid or fluidized material may include such agents as acid-acting substances, basic-acting substances, salts, reducing agents, oxidizing agents, halogenating agents, solvents of all kinds, solutizing agents, neutral or reactive absorbents, adsorbents such as clay, charcoal, gels, cation and anion exchange resins as well as such agents commonly employed in the refining, separating, extracting, concentrating and purifying of fluid type mixtures.

The treatment of the materials under consideration, for the purposes of this

BEST AVAILABLE COPY

invention, is undertaken in an annular zone formed between two continuous surfaces of revolution, one being inside the other, which surfaces do not touch each other, wherein either one or both of said surfaces is rotated about its axis within a predetermined range of rotational velocities depending upon the properties of the materials being contacted and the particular design and operation of the contacting apparatus.

The annular zone previously referred to and in which contact or treatment of the materials takes place, is a space formed between the outer curved surface of an inner surface of revolution and the inner curved surface of an outer surface of revolution. The axes of these two surfaces need not be coincident. There may be more than two surfaces of revolution, such as a series of surfaces of revolution forming for example several concentric annuli. In a preferred and a specific case, involving only two surfaces of revolution about a common axis, the annular zone may be defined as being bounded by two surfaces which are formed by rotating two lines (straight or curved) about a straight line or axis, all three of which lines meet the following conditions: (1) lie in the same plane; (2) have a definite length extending between two boundaries defined by two parallel planes perpendicular to said straight line, and (3) do not cross each other within said definite length. For example, all three lines may be straight lines, such as to form a zone comprising a truncated hollow cone, a hollow cylinder or a combination of one or more of these two figures positioned adjacent each other along the same axis.

The two surfaces of revolution may be relatively rough, smooth, or polished, as desired, and are preferably positioned so that their common axis is not in a horizontal position, i.e. one end should be substantially higher than the other.

The above mentioned differently shaped contact surfaces and annular zones permit varying peripheral velocities of the surfaces and provide varying annular cross-sectional areas from one end of the zone to the other. These zones are applicable for contacting fluid systems which change in processing in any one or more of the following properties: volume, weight, viscosity, surface tension, ratio of the phases, etc., as they pass through the zone. Such changes may be due to the addition and/or withdrawal of fluid at one or more points along the column or due to the physical or chemical effects that occur between two fluids as they continue in contact through the zone. It is

desirable, however, that these changes in shape of the surfaces should not be abrupt. For systems in which the above variables change but little, the most desirable form is that of two concentric cylinders.

Regarding the physical proportions of the apparatus, it is desirable that the annular contact zone be at least twice as long as the internal diameter of the outer surface and it may be as long as operational and mechanical limitations permit. The length of the zone should be at least about four times, and preferably at least about 10 times, the minimum annular clearance between the two surfaces of said zone. The longer the column is, the more theoretical stages it contains, and the lower is the loss resulting from "end effect" at feed inlets and at the ends of the zone, where disorderly distribution is initially prevalent. The ratio of the width of annular space to the diameter of the inner surface should be not more than about 5:1 and not less than about 1:50 and preferably between 2:1 and 1:10.

In the operation of an apparatus embodying one of the above-mentioned types of annular zones, it is essential that there be obtained a difference in rotational absolute angular velocity between any two adjacent contact surfaces. For example, one surface may be at rest or both surfaces may rotate in either the same or in opposite directions. In many systems it is preferred that the inner surface rotate at a greater peripheral speed than the outer surface, and the simplest and most preferred embodiment is that of an inner rotating cylindrical surface and a stationary outer cylindrical surface. By varying the rate of rotation of one surface of revolution relative to the other, it is possible to control the efficiency of contact and to diminish the tendency of the two phases to form emulsions.

Further, in the operation of an apparatus embodying one of the above-mentioned types of zones, it is essential that while the fluids pass through the zone they contact both surfaces at all times. These fluids may be passed through the zone either concurrently or counter-currently and either fluid may be fed continuously or intermittently into the zone. In the case of concurrent flow, it is not necessary that the two phases possess different densities; however, for countercurrent flow the converse is essential. Therefore, in the latter case, a density difference of more than about .02 gms./cc. between the two phases is desirable.

For a given fluid system, the extent of contact in a rotary contactor is determined by the extent to which the dispersed phase is carried by vertices formed in the continuous phase in the annular contact zone. The extent of contact in a separation process may be defined as the rate of transfer of one or more components from one phase to another phase per unit time per unit volume of fluid mixture in the contact zone.

In the operation of a rotary contactor apparatus of the type disclosed, when the annular zone is filled with a fluid, there is a shearing force set up in the fluid which produces a particular type of flow in the annular zone. In the apparatus of the type having two concentric cylindrical surfaces of revolution, wherein the inner surface rotates and is hereinafter called the "rotor" and the outer cylinder is stationary, hereinafter called the "stator," the normal flow is in and around a series of paired annular vertices appearing as "double doughnuts" (or *terti*) stacked axially, one on top of the other around the rotor. The flow of the fluid in these vertices is, firstly, around the annulus or around the rotor, and secondly, around the periphery of the "doughnut" between the rotor and the stator, as in a smoke ring, and in such directions that any two sections of adjacent "doughnuts" rotate as though they were geared together. The width of the vertex in most cases corresponds to the distance between the rotor and stator, so that the height of the "double doughnut" is at least about twice this distance.

Upon introducing another fluid substance into the continuous fluid in the contact zone, which substance forms a separate phase with the fluid in the zone, dispersion of the substance occurs. The dispersed particles are, more or less, carried in the paths of flow of the continuous phase in and around these vertices. The degree of dispersion and the degree to which the dispersed phase is carried in these vertices, as well as the rate of transfer from one phase to the other, depend upon (1) the magnitude of the shearing force in the continuous phase and (2) the relative properties of the continuous phase and dispersed phase. These factors are:

1. The relative angular velocity of rotation of adjacent contact surfaces.
2. The radii of the contact surfaces.
3. The rate of flow of the phases (continuous and dispersed) through the contacting zone.
4. The settling force either natural, that is by gravity, or induced, such as by

electrostatic or magnetic fields.

5. The roughness of the contactor surface.

6. The temperature.

7. The pressure.

8. The dynamic stability of the apparatus.

9. The viscosity of the phases.

10. The density of the phases and the density differential between them.

11. The interfacial tension between the phases.

12. The adhesion tensions between the phases and cylindrical surfaces.

13. The film coefficients of mass transfer between the phases.

14. The distribution of the distributed component between the phases.

15. The pH of the system.

The independent factors which affect the rate of transfer of two liquid phases in countercurrent flow for a given column and a given liquid system are: the speed of rotation, the rate of flow of each phase into the zone, the phase which is dispersed, the temperature, and the pressure.

The relative size of the particles of dispersed phase to the size of the vertex is another factor in determining the degree to which the particles are carried by the vertex. Thus in contactors having the same rotor diameter to stator diameter ratio, particles of a given size are more easily carried in vertices in larger contactors than they would be in smaller contactors.

When the substance to be dispersed is a liquid or a gas, the shearing forces in the continuous phase cause it to be broken up into particles of a limited size distribution, which size depends also upon the relative properties of the two phases. This means that the particles of the dispersed phase are sufficiently uniform in size and shape so that about 90% of the dispersed phase is in particles having sedimentation velocities which lie within the range from about $\frac{1}{4}$ to 4 times the average sedimentation velocity (the average sedimentation velocity,

$$V = \frac{F_d}{H} + \frac{F_c}{1-H}$$

), wherein H is the holdup or volume fraction of the dispersed phase in the contact zone in the zone, and F_c and F_d are the feed rates of the continuous and dispersed phase in volume per unit area per unit time, respectively. The particles of the dispersed phase remain at substantially the same size throughout their passage through the contact zone.

The formation of particles of uniform size has the advantage of preventing

emulsification and/or foaming often caused by the formation of at least some exceedingly small sized particles in other types of contacting apparatus.

- 5 Accordingly, the process of this invention is particularly applicable in contacting fluid systems which, in other apparatus, would give rise to emulsification difficulties.
- 10 As the relative difference in angular velocity between the two surfaces of the contact zone is increased, more and more particles of the dispersed phase are carried by the vertices. Different amounts
- 15 of particles carried by the vertices of the continuous phase form different visual patterns which are a visual indication of the extent of contact between the dis-

persed phase and the continuous phase. Therefore, the pattern can be used as a 20 way of estimating the degree of contact between the two phases.

There is a definite range of relative angular velocities within which efficient contacting is obtained and within which 25 definite patterns are observed. This range of efficient contacting is the range of "orderly distribution," and particularly the region in this range of the "herringbone pattern." The regions and 30 corresponding patterns, which are obtained as the relative angular velocity between the surfaces of the contact zone increases from zero up to and beyond the range of efficient contacting, may be 35 described as follows:

Region	Apparent Size of the Dispersed Phase	Apparent Distribution of the Dispersed Particles in the Contact Zone	Flow of Apparent Pattern of the Particles through the Annular Contact Zone	See Figures	Efficiency of Contact between the Dispersed and Continuous Phases
A	Streamers and large particles	Irregular	Helical	VIIa, b, c	Poor
B	Smaller particles and of more uniform size	Orderly distribution	From slightly pitched to horizontal	VIIIa VIIIb	Fair
C	Small uniformly sized particles	Orderly distribution	Horizontal around the rotor; particles concentrated in the vortices	IXa IXb	Good
D	Small uniformly sized particles	Orderly distribution	Around the rotor in the vortices and around the sections of the vortices	Xa Xb	Excellent
E	Small uniformly sized particles	Orderly distribution	Around the rotor in the vortices and around the sections of the vortices	XIa XIb	Excellent
F	Small particles	Random turbulence	None, random		No controlled flow through the contact zone

A change in the interfacial tension between the phases has a definite effect on the appearance of the particles (shape and size) in any given apparatus at any given rotor speed, but this effect does not destroy the general pattern, as indicated in the table above.

With certain rotary contactors and certain fluid systems the "doughnut" type vortices are replaced by a double helix type pattern appearing like a rotating "barber pole." In the double helix the particles move around the rotor and around the periphery of a section of the helix between the rotor and the stator in such directions that two adjacent helix sections rotate as though they were geared together. The double helix may first appear in any one of the Regions C, D or E of "orderly distribution."

In Region F, random turbulence results in failure of the dispersed phase to travel independently in the direction of its gravitational force. In a counter-current system, this causes the column to "flood," that is, both phases are simultaneously withdrawn from both ends of the zone. At the point of random turbulence the desired types of contact patterns disappear, and the advantages, such as orderly flow, orderly distribution, minimum emulsifying tendencies, controlled residence times, are no longer obtained.

The point at which flooding occurs may be determined as follows:

Plot on the same graph (not shown) the two curves defined by (1) the ratio of the volume of dispersed phase to the volume of continuous phase in the column against the peripheral speed of the rotor, and (2) the ratio of the linear velocities of the two phases through the column against the peripheral speed of the rotor. The point of intersection of curves (1) and (2) defines the point of flooding. The phenomenon of flooding may be represented by the following equation:

$$H = \frac{1}{\left(\frac{F_o}{F_d}\right)^{\frac{1}{2}} + 1}$$

wherein H is the holdup or volume fraction of the dispersed phase in the contact zone in the column when flooding occurs, and F_o and F_d are respectively the feed rates of the continuous and dispersed phases, expressed in volume per unit area per unit of time.

Another criterion for efficient contact in the rotary contactor of this invention may be expressed by a Reynolds number

for rotational flow of the fluid mixture in the annular zone.

When only the inner surface of revolution of the annular zone is rotated, the product of the peripheral velocity times the clearance between the two surfaces at any section through the zone divided by the kinematic viscosity (the viscosity divided by the density) of the fluid mixture in the annular zone should be greater than about 2500 and preferably greater than about 3000; these values represent dimensionless numbers.

When only the outer surface of revolution of the annular zone is rotated a preferred condition of operation is when the product of the peripheral velocity times the clearance between the two surfaces of revolution at any section through the zone divided by the kinematic viscosity of the fluid mixture in the zone is greater than about 2000, a dimensionless number. The critical value under these conditions is less than in the preceding instance, because stable eddies are maintained over a wider range in this type of apparatus.

In an apparatus of the type having an "inner rotor and outer stator" when contacting aqueous systems wherein the kinematic viscosity of the fluid mixture in the column is not substantially greater than that of water, effective contact is obtained when the peripheral velocity of the inner cylinder is greater than about 2.5 or 3 feet per second.

In a contactor apparatus where both surfaces of revolution are rotated in the same direction, the ratio of the angular velocity of the inner surface of revolution to the outer surface of revolution should be greater than the square of the ratio of the radius of said outer surface to the radius of said inner surface, that is, at any section through the contact zone.

The operating range of the rotary contactor for a given efficiency is very broad in that both the rotor speed and the total throughput may be varied within comparatively wide limits. This is an important advantage in industrial processes. Thus, when driving the rotor with a variable speed motor many different effects may be easily obtained, and the efficiency of the contacting may be controlled at will within wide limits. Changing the relative angular velocity of the surfaces in a given column is analogous to changing the type or size of the packing material, such as Raschig rings, saddles, etc., in a packed tower; or the size of the orifice jets in a spray tower; or the hole size in a perforated plate tower. The ease of changing the speed is in marked contrast to the difficulty in changing the

packing, jets or hole size in other columns.

Still another important advantage of the rotary contactor of this invention is the ease of regulating residence time of the dispersed phase in the contact zone by controlling the relative peripheral velocity between the surfaces of the zone. This is particularly helpful in effecting the orderly procedure of all the dispersed particles through the zone.

In considering the effect of variations of the size of the rotary contactor or its efficiency in comparison with similar variations of other contacting devices such as packed or sieve plate towers, it is believed that with increasing size the rotary contactor is progressively more efficient than the packed or sieve plate tower. Also, the initial efficiency of a rotary contactor is believed to be materially greater than that of a packed tower of the same size.

Rotary contactors are definitely advantageous compared to other types of contactors when the former have effective annular cross-sectional areas of more than about 1.5 square inches.

The lower the interfacial tension between the two phases, the easier it is to disperse one phase in the other. Therefore, surface-active agents may be added to either or both fluid phases to reduce the interfacial tension in the contact zone. Within limits, surface-active agents may also be used in this manner to prevent the formation of emulsions in the apparatus. The proper choice of a surface-active agent, either cation-active or anion-active, or both depends upon the nature of the fluids being contacted. Some suitable surface-active agents are organic compounds containing one or more of the following radicals: metal or acid sulfate, sulfonate, phosphate, phosphonate, or carboxylate; hydroxyl or sulfhydryl, carbonyl, formyl, ether, or ester; primary, secondary, or tertiary amine, or quaternary ammonium and other onium base radicals; cyano, thiocyno or nitro; halide. For example, it is often desirable to use such agents in the extraction of penicillin from aqueous solutions with solvents to prevent emulsification of the impurities in the penicillin solution.

Some illustrative uses for this invention are:

I. GAS-LIQUID AND LIQUID-GAS SYSTEMS.

General gas purification processes, gas stripping, including refluxing in distillation and extractive distillation, gas absorption, gas humidification and dehumidification.

II. GAS-SOLID SYSTEMS (wherein the solid is a powdered fluidized solid).

Adsorption operations such as the separation of methane and other light gases from ethane and ethylene in natural gas by adsorption on charcoal; adsorption reactions employing silica gel and other powdered adsorbent materials; fractionation of powder by elutriation or gas flotation. Chemi-adsorption operation such as the adsorption of olefins, diolefins or acetylenes by cuprous salts.

III. LIQUID-LIQUID SYSTEMS.

Solvent extraction processes for the separation of liquid mixtures containing organic and/or inorganic components of different degrees of polarity and/or saturation or molecular configuration such as the separation of olefins, diolefins, acetylenes, naphthenes, aromatics, alkyl phenols, thiophenols, nitrogen bases, etc. from petroleum or coal tar oil fractions as well as from their synthetic mixtures with one or more selective solvents such as sulfur dioxide, furfural, phenols, sulfonates, copper-containing solutions, aldehydes, ketones, ethers, esters, amines, nitrites, nitrohydrocarbons, and the like; the refining and purification of fatty acids, fatty oils, animal and vegetable oils and essential oils such as tung oil, linseed oil, soy bean oil, lemon oil, resin oil, tall oil, cottonseed oil, and the like with suitable solvents such as ethanol-amine, furfural sulfonate and the like; purification of dialkyl amine such as diisopropylamine by its separation from the corresponding monoalkyl amine, other amines and other contaminants with a hydrocarbon solvent; the extraction, concentration and purification of antibiotics such as penicillin with selective solvents; the separation of polymers with solvents; the separation of epi- and di-chlorohydrins from water with diisopropyl ether or other related solvent; and many other solvent extraction processes, particularly those wherein a large number of extraction stages is required because of small differences in solubilities of the two substances to be separated in the solvent employed for their separation. Neutralization reactions including liquid caustic alkali treating processes such as the treatment of hydrocarbon oils containing acidic compounds such as hydrogen sulfide, mercaptans including thiophenols, alkyl phenols, naphthenic acids, sulfonic acids, by contact with aqueous caustic alkali solutions including sulfonate solutions and the like; the separation of alkyl phenols from thiophenols and the separation of mixtures of alkyl phenols or of thiophenols into their com-

ponents; the separation and purification of mixtures of sulfonic or naphthenic acids into their components. Acid treating processes such as the treatment of mineral oils with sulfuric acid to absorb gum forming components, and other impurities are also included.

IV. LIQUID-SOLID SYSTEMS.

Leaching reactions such as the activation of clays with acid, the washing of precipitates, the deoiling of wax with solvents, the regeneration of adsorbents, the deoiling of adsorbents, the extraction of oil from shales, the extraction of oils from asphalts, the extraction of fatty oils from their meals or flakes including cotton seeds, soy bean flakes, with organic solvents. The separation of proteins, including antigens, allergenic proteins, and other blood components, of hormones, vitamins, and antibiotics from solutions containing them by adsorbing them on a solid adsorbent material. The separation of compounds of different degrees of polarity or saturation such as phenols from hydrocarbons, aromatics from non-aromatics, by absorption on a solid absorbing agent such as silica gel. Also included herein are processes involving ion exchange substances including ion exchange resins, Permutite type substances.

V. LIQUID-GAS-SOLID SYSTEMS.

Mineral separation processes such as froth flotation processes.

VI. LIQUID-LIQUID-SOLID SYSTEMS.

The separation of proteins, including antigens, antibodies, allergenic proteins, and other blood components, of hormones, vitamins and antibiotics from mixtures containing them by dissolving the mixture in a continuous phase and dispersing therein both adsorbing particles and an

immiscible liquid phase whereby the adsorbing particles would be carried at the interface between the two liquid phases.

VII. LIQUID-LIQUID-LIQUID SYSTEMS.

Transfer of a solute from one solvent to a second solvent by contacting them countercurrently in a dispersing medium, such as extracting phenols from a high boiling hydrocarbon solvent into a low boiling denser solvent by contacting them countercurrently in an aqueous alkaline dispersing solution.

The following examples illustrate the effectiveness of several contacting processes in the apparatus by the method of this invention.

EXAMPLE I.

Gasolines having an ASTM 5% boiling point at 135° F. and a 90% boiling point at 338° F. and containing different amounts of tertiary-butyl mercaptan were countercurrently contacted with a 3N aqueous solution of potassium hydroxide in two different sized glass contactor apparatus similar to that shown in Figure V. The apparatus of Runs 1 and 2 had an effective column height of 25 inches, a stator diameter of 1.312 inches, and a rotor diameter of 1 inch, while the apparatus of Runs 3 and 4 had a column height of 25 inches, a stator diameter of 2.185 inches and a rotor diameter of 1.476 inches. The runs were made with the following (see Table A below) different throughputs, different rotor speeds, and following degrees of extraction were obtained from which the efficiency in theoretical stages per foot of length was calculated according to the method described in the article by Hunter and Nash supra. The caustic phase was the continuous phase and the gasoline phase formed the dispersed phase.

TABLE A.

Run No.	Throughput of KOH	Gals./in. ² / min. of Gasoline	% tert.-butyl mercaptan in feed as S%	% tert.-butyl mercaptan in raffinate	Peripheral Velocity ft./sec.	Partition Coefficient of KOH to Gasoline	Temp. (° C.) of Contact	Efficiency in stages per ft.
1	.0040	.0195	.046	.0216	2.14	6.44	23.3	.422
2	.0039	.0096	.0470	.000432	3.30	6.53	23.0	5.61
3	.0011	.0054	.0479	.00817	4.65	6.36	23.8	1.67
4	.0018	.0088	.0500	.00169	4.65	6.53	23.0	3.45

Runs 1 and 2 show that with otherwise constant conditions an increase in peripheral speed to or above the neighborhood of 2.5—3 ft./sec. leads to a disproportionate increase in efficiency, while Runs 3 and 4 illustrate that where the peripheral speed is constant and the total throughput is increased by about 50% the efficiency is increased by slightly more than 100%.

A definite "herringbone" pattern was observed in the contactor during each of the above runs, except that of Run 1 which was operated at a peripheral speed below that required to obtain a pattern.

EXAMPLE II.

A narrow boiling fraction of petroleum alkyl phenols had an initial boiling point of 195.5° C., a final boiling point of 206.5° C., a 5% distillation temperature of 197° C., a 50% distillation temperature of 199° C., and a 95% distillation temperature of 205° C. An acetylation test shows that the mixture corresponds to about 57% xylenols and ethyl phenols (comprising *o*-ethyl phenol and 2,6-, 2,4-, and 2,5-xylenols) and 43% of cresols (comprising substantial amounts of all three cresols). This mixture was dissolved in a sufficient amount of equal volumes of iso-octane and benzene to give a solution containing 200 grams of alkyl phenols per liter of solution. The resulting solution was extracted counter-currently with aqueous sodium hydroxide solution containing 2 moles per liter of NaOH in a rotary contactor similar to that shown in Figure V and having a rotor 25 inches long, a rotor diameter of 1.476 inches and a stator diameter of 2.185 inches. The solution of alkyl phenols was introduced at the rate of .01125 gal./in.²/minute and the caustic soda solution at the rate of .0053 gal./in.²/minute, the latter being the continuous phase. The rotor speed varied from 3.22 to 3.65 ft./sec. Well developed "herringbone" patterns were present under these operating conditions.

The xlenol and ethyl phenol content of the raffinate was 96% and of the extract was 7.9%.

EXAMPLE III.

Several aqueous penicillin solutions were extracted with different solvents in a rotary contactor of the type shown in Figure V having a stator diameter of 2.0 inches, a rotor diameter of 1.3 inches, and a length of 41 inches. The column was operated at between about 0° and 3° C. to prevent decomposition of the penicillin in the solution. The following data show the recovery and purity of penicillin obtained from different solutions under the following conditions:

TABLE B.

Run No.	Purity of Penicillin in aqueous feed solution Oxford Units/mg. ^a of solids	Solvent	Continuous Phase	Feed Ratio of Continuous: Dispersed Phase	Peripheral Velocity in ft./sec.	% Recovery of Penicillin in Extract	Purity of Penicillin in Raffinate	Purity of Penicillin in Oxford Units/mg. Extract
1	175	MIBK	MIBK	1:1	5.68	97.5%	35	810
2	71	MIBK	Aqueous feed	1:1	3.69	92.1%	71	1043
3		50-50% CHCl ₃ -MIBK	CHCl ₃ -MIBK	0.67:1	5.68	99.5%		
4		CHCl ₃	CHCl ₃	0.67:1	6.2	95%		
5		CHCl ₃	Aqueous feed	0.67:1	3.12	96%		

^aSee article by Florey and Jennings in British Journal of Experimental Pathology, Vol. 23, p. 120, of June 1942.

A definite "herringbone" pattern was observed in the contactor during each of the above runs. Runs 4 and 5 in the above Table B show that efficient extraction can be obtained with two liquids of relatively great density difference (chloroform=1.5, water=1.0) when either phase is continuous.

EXAMPLE IV.

Two methyl isobutyl ketone solutions of acetic acid were contacted with water

in a contactor with a glass stator similar to that shown in Figure V at about room temperature at different rotor speeds and at different water-to-feed ratios. The percent of the acetic acid extracted by the water and the efficiency of the contactor in the number of theoretical stages per foot, were determined for each run. Data on the two representative runs are shown in Table C below. The efficiencies were calculated according to the article mentioned in Example I above.

TABLE C.

Run No.	Dimensions of Column in inches		Concentration of acetic acid in feed moles/liter	% Acetic acid in Raffinate moles/liter	Acetic Acid in Extract moles/liter	Total Throughput gal./in. ² /min.	Feed Ratio		Peripheral vel. in ft./sec.	Efficiency stages/ft.
	Stator diam.	Rotor diam.					Continuous	Dispersed		
1	8.5	6.25	20	.526	0.091	.576	.065	1.32:1	10.2	1.41
2	8.5	6.25	20	0.466	0.084	0.518	.185	1.36:1	8.16	1.39

These examples indicate the flexibility of a rotary contactor and show that equal efficiency is obtained by change in peripheral velocity with a converse change in through-put.

A definite "herringbone" pattern was observed in the contact zone during each of the above runs in Table C.

EXAMPLE V.

Two different crude lubricating oil bulk distillates having viscosity indices below about 50 were contacted with furfural in a glass apparatus similar to that shown

in Figure V having a stator diameter of 3.75 inches, a rotor diameter of 2.14 inches, and 20 inches between feed ports along the column. The rotor was operated for each run at 300 R.P.M. The furfural was heated and introduced at the top of the column and the lubricating oil was heated and introduced at the bottom of the column. The raffinate phase of extracted oil was removed from the top of the column, freed of furfural, and tested for viscosity index (V.I.). The following table shows the conditions and results of each run:

Run No.	Lubricating Oil		Throughput in		Column Temp. ° C.		Raffinate	
	S.U. sec. at 210° F.		cc./min. Lube Oil	Furfural	Top	Bottom	V.I.	% Yield
5 1	200		640	700	88	79	55	50
2	60		650	1075	96	69	44	41

In each of the runs in the above table the lube oil was the dispersed phase.

A "herringbone" pattern was not only observed in these runs, but also in runs where the furfural was the dispersed phase.

EXAMPLE VI.

A gaseous mixture of 27.1 vol. % H_2S and remainder of nitrogen was contacted countercurrently with an aqueous solvent solution containing 15.1 wt. % of diethanolamine at atmospheric pressure and room temperature in a glass rotary contactor having a rotor diameter of 0.55 inches, a stator diameter of 0.75 inches and an effective length of liquid therein

of 28.0 inches. The rotor was rotated at a speed of 1395 R.P.M., the solvent was fed in at the top at a rate of .074 gal./hr. 25 and the gaseous mixture was fed in at the bottom at a rate of .425 cu. ft./hr. All of the H_2S was separated from the N_2 .

Comparative runs were made with the 30 same gaseous mixture but with an aqueous solvent solution containing only 6 wt. % diethanolamine. The rotary contactor employed had a glass stator 0.87 inches in diameter and a metal rotor of 0.63 35 inches in diameter. The following conditions and results of the runs were:

	Rotor Speed	Liq. Ht. in inches	Inlet gas rate ft. ³ /in.	Solvent Rate Gal./hr.	H_2S conc. in inlet gas % v.	% H_2S Removed
40	1240	33.0	.398	.039	24.5	72.5
	0	32.0	.398	.036	29.7	56.5

Orderly distribution of the dispersed phase was obtained when the rotor was rotated both at 1240 and 1395 R.P.M.

EXAMPLE VII.

A cracked light hydrocarbon gas boiling below the butane-butylene fraction and containing 2.8 vol. % acidics, 25.2 vol. % H_2 , and 10 vol. % products soluble in 98% H_2SO_4 containing 3% Ag_2SO_4 was concurrently contacted at atmospheric pressure and room temperature with powdered carbon black in a rotary contactor having a stainless steel rotor .63 inches in diameter, a glass stator 0.82 inches in diameter and an

effective contact length of 26 inches. The rotor was operated at 2200 R.P.M., and the gas was fed into the contact zone at a rate of 120 gms./hr. The effluent gas 60 contained 34.1 vol. % H_2 , .6 vol. % acidics, and 0.0% solubles in said acid. A definite "herringbone" pattern of the carbon particles was observed.

Dated the 30th day of July, 1945.

ELKINGTON & FIFE,
Consulting Chemists & Chartered Patent Agents,
Bank Chambers,
329, High Holborn, London, W.C.1,
Agents for the Applicant.

COMPLETE SPECIFICATION

Improvements in Method of Contacting Fluids One with Another

I, JAMES GEORGE FIFE, M.Sc., Ph.D., F.R.I.C., Consulting Chemist and Chartered Patent Agent, of Bank Chambers, 329, High Holborn, London, W.C.1, a British Subject, do hereby declare the 65 nature of this invention and in what 70

manner the same is to be performed, (as communicated to me by Shell Development Company, a corporation organised under the laws of the State of Delaware, United States of America, of 100, Busa 75 Street, San Francisco, California, United

States of America), to be particularly described and ascertained in and by the following statement:—

The present invention relates to methods of contacting fluids and fluidized materials. More particularly, the present invention pertains to the treatment of mixtures of fluids in which advantage is taken of the formation of discrete phases.

It is an object of the invention to provide a method for the efficient contacting of two or more substances which form at least two phases in contact with each other, at least one of which substances is fluid.

This and other objects are within the purview of the present invention and will be elaborated upon in more specific portions of the specification.

The fluid or fluidized material, hereinafter called "fluids" includes liquids, gases, vapors, powders and finely divided bodies, which are capable of forming a multiphase system or, more specifically, a dispersion of at least one type of material in a continuous phase of the other, which phases remain discrete throughout the major portion of the contact zone. Illustrative of the general types of heterogeneous systems contemplated are: gas-solid, gas-liquid, liquid-gas, liquid-liquid, liquid-solid, liquid-liquid-solid, liquid-gas-solid, gas-liquid-solid, liquid-liquid-gas, liquid-liquid-liquid, and gas-liquid-liquid systems wherein the first-named constituent comprises a continuous phase and the second constituent (with the third) comprises a dispersed phase or phases. Each phase may comprise a plurality of components such as a mixture of solvents of different capacities or different degrees of selectivity, mixtures containing diluents for reduction of viscosity, for change in surface tension or interfacial tension, or for alteration of internal density. The fluid or fluidized material may include such agents as acid-acting substances, basic-acting substances, salts, reducing agents, oxidising agents, halogenating agents, solvents of all kinds, solutizing agents, neutral or reactive absorbents, adsorbents such as clay, charcoal, gels, cation and anion exchange resins as well as such agents commonly employed in the refining, separating, extracting, concentrating and purifying of fluid type mixtures.

It has been found, according to this invention, that efficient contacting of two or more substances which form at least two phases in contact with each other, at least one of which substances is a fluid, may be effected by a method

which comprises contacting said substances by passing said phases simultaneously, and preferably in counter current, into an annular zone formed between two surfaces of revolution, one inside the other and out of contact with each other, wherein one of said surfaces rotates about its axis at a different angular velocity from that of the other surface sufficient to maintain one phase dispersed in the other (continuous) phase and to maintain an orderly distribution, as hereinafter defined, of the dispersed phase in the continuous phase.

It has been found that the operating conditions which produce the above orderly distribution lead to efficient contacting, and that a criterion of orderly distribution as opposed to random turbulence may be established by visual observation. Orderly distribution has been found to be associated with regular visual patterns of the dispersed phase in the continuous phase in the annular zone between the two surfaces, and in particular with herringbone patterns as discussed more fully hereinafter, whereas random turbulence has been found to be associated with no regular visual pattern in the two phases.

The term "orderly distribution" as used herein is therefore to be understood as referring to a condition associated with regular visual patterns of the dispersed phase in the continuous phase.

To illustrate the various annular contact zones in which the treatments or contacts are to be effected and to illustrate other physical aspects of our invention, reference is made to the accompanying drawings, wherein:

Figures 1a to IVd inclusive depict vertical partial sections of different shaped annular zones. Figure V is a vertical partial section of a contact apparatus embodying the contact zone shown in Figure 1a. Figures VI and XIV illustrate different paths of flow for a fluid in a contact zone of the type shown in Figure V. Figures VIIa to XIIIb inclusive are drawings of patterns formed by the dispersed phase in a contact zone of the type shown in Figure V. Figure XV shows a family of curves plotted at different throughputs against extraction efficiencies and against different rotor speeds. Figure XVI shows a family of curves plotted for different sized rotary contactors against different throughputs and different optimum extraction efficiencies. Figure XVII shows two curves comparing efficiency with size of the apparatus for the present rotary contactor apparatus and a packed tower.

The annular zone previously referred

to in which contact or treatment of materials takes place is the zone formed between the outer curved surface of an inner surface-of-revolution and the inner curved surface of an outer surface-of-revolution as illustrated by Figures Ia to IVd inclusive. The axes of these two surfaces need not be coincident (see Figure Ic). There may be more than two surfaces-of-revolution forming several concentric annuli (see Figure Ib).

The two surfaces-of-revolution may be relatively rough, smooth or polished, as desired, and are preferably positioned so that their common axis is not in a horizontal position, i.e. one end should be substantially higher than the other. Other configurations and constructions of the annular zone are self-evident from the drawing.

The above differently shaped contact surfaces and annular zones permit varying peripheral velocities of the surfaces and provide varying annular cross-sectional areas from one end of the column to the other. These zones are applicable for contacting fluid systems which change in processing in any one or more of the following properties: volume, weight, viscosity, surface tension and ratio of the phases, as they pass through the column. Such changes may be due to the addition and/or withdrawal of fluid at one or more points along the column or due to the physical or chemical effects that occur between two fluids as they continue in contact through the zone. It is desirable, however, that these changes in shape of the surfaces be abrupt. For systems in which the above variables change but little, the most desirable form is that of two concentric cylinders.

Regarding the physical proportions of the apparatus, it is desirable that the annular contact zone be at least twice as long as the internal diameter of the outer surface and it may be as long as operational and mechanical limitations permit. The length of the zone should be at least about four times, and preferably at least about ten times, the minimum annular clearance between the two surfaces of said zone. The longer the column is, the more theoretical stages it contains, and the lower is the loss resulting from "end effect" at feed inlets and at ends of zone, where disorderly distribution is initially prevalent. The efficiency of a column is expressed as the number of theoretical stages per foot of length of the column calculated according to the method described in the article by Hunter and Nash in the Journal of the Society of Chemical Industry, Vol. 51, p. 285

T(1932). The ratio of the width of annular space to the diameter of the inner surface should be not more than about 5:1 and not less than about 1:50 and preferably between 2:1 and 1:10.

In the operation of an apparatus embodying one of the above-mentioned types of annular zones, it is essential that there be obtained a difference in rotational absolute angular velocity between any two adjacent contact surfaces. For example, one surface may be at rest or both surfaces may rotate in either the same or in opposite direction. In many systems it is preferred that the inner surface rotate at a greater peripheral speed than the outer surface, and the simplest and most preferred embodiment is that of an inner rotating cylindrical surface and a stationary outer cylindrical surface. By varying the rate of rotation of one surface of revolution relative to the other, it is possible to control the efficiency of contact and to diminish the tendency of the two phases to form emulsions.

Further, in the operation of an apparatus embodying one of the above-mentioned types of zones it is essential that while the fluids pass through the zone they contact both surfaces at all times. These fluids may be passed through the zone either concurrently or countercurrently and either fluid may be fed continuously or intermittently into the zone. In the case of concurrent flow, it is not necessary that the two phases possess different densities; however, for countercurrent flow the converse is essential. Therefore, in the latter case, a density difference of above about .02 gms./cc. between the two phases is desirable.

For the purpose of illustration, the rotary contactor in Figure V will now be referred to as operating as a liquid-liquid solvent extractor, wherein a liquid mixture to be separated is the lighter and continuous phase and flows upwardly through the annular cylindrical zone countercurrently to a solvent which is the heavier and dispersed phase. The rotary contactor comprises, in this case, an outer cylindrical surface 1, called the "stator," and an inner cylindrical rotating surface 2, called the "rotor." The rotor is mounted at each end in bearings 3 and 4 and rotated by a variable speed motor 5. Above and below the heavy phase and light phase inlets, 6 and 7 respectively, are provided quiescent zones which are shown here to be settlers 8 and 9 through which the light phase and heavy phase products are separately withdrawn from lines 10 and 11, respec-

tively. Each settler is provided with a return or recycle line 12 and 13 respectively, for settled carryover, and with inlet lines 14 and 15, respectively.

6 The light phase introduced through line 7 passes up and around the annular cylindrical surface between the stator 1 and rotor 2 and is withdrawn through outlet line 10. The heavy phase introduced through line 6 is dispersed in the continuous light phase in the contactor by the action of the rotor. The dispersed phase then passes down through the annular cylindrical zone countercurrently to the continuous phase in a manner more fully described later, and is coalesced and withdrawn through lower outlet line 11.

20 An interface level between the two phases is maintained in either top or bottom settler 8 or 9 by suitable level control means such as electric controllers 38 or 39 at the desired location of the interface. This means that elements 38 or 39 control valve 40 for the withdrawal of the heavy phase through line 11. The location of the interface will depend upon which of the two phases is the continuous and which is the discontinuous phase. The rule is that the discontinuous phase travels toward the interface level. Thus if the light phase is the dispersed phase, the interface level will be in the top settler 8; while if the heavy phase is the dispersed phase, the interface level will be in the bottom settler 9. Usually the dispersed phase passes through the contact zone more rapidly than the continuous phase and if one of the materials to be contacted is relatively unstable, it is usually desirable that it be the dispersed phase.

45 Many modifications of the described apparatus are possible without departing from the scope of this invention; for example, the stator chamber may be closed at the top and the apparatus may be operated under a reduced or increased pressure. The heavy and/or light phase inlets 6 and 7, instead of being placed as shown, may join recycle lines 12 and 13, respectively, or be placed elsewhere along the column; the column may have side inlet lines where the same or different fluids may be added (such as for controlling pH of fluid systems); or it may have side draw-off lines for separating side streams which may be removed, or returned to the system in part or in toto at points along the column. The settler may comprise any means known in the art to enhance settling other than the long tubes 14 and 15 shown in Figure V. The rotor may be supported solely by a top bearing or by a bottom bearing. The apparatus may be

jacketed and fluids may be circulated through the jacket and/or the rotor to control and maintain any desired temperature. The motor 5 may be mounted to drive either end or both ends of the rotor either directly or indirectly. If the rotor is relatively long in comparison to its diameter, it may be steadied by bearings between its ends. The rotor may be driven by any suitable power source. Independent extraneous forces such as sonic and ultra-sonic vibrations, ultraviolet and infra red light, electromagnetic, electrostatic, gravitational fields, or heat, may be imposed upon the contents of the annular zone during the contacting period.

The apparatus may be made of any suitable material which will withstand the strain of the rotational and other forces present during its operation and not react with the heterogeneous fluids being contacted therein. Some suitable materials are various steels including stainless steel, nickel, brass, Monel metal (Monel being a Registered Trade Mark), aluminum, magnesium, glass, earthenware, wood, and plastic, depending upon the nature of the fluids contacted and the temperature and pressures employed.

95 For a given fluid system, the extent of contact in a rotary contactor is determined by the extent to which the dispersed phase is carried by vortices formed in the continuous phase in the annular contact zone. The extent of contact in a separation process may be defined as the rate of transfer of one or more components from one phase to another phase per unit time per unit volume of fluid mixture in the contact zone.

In the operation of a rotary contactor apparatus of the type disclosed, when the annular zone is filled with a fluid, there is a shearing force set up in the fluid which produces a particular type of flow in the annular zone. In an apparatus of the type shown in Figure V, the normal flow is in and around a series of paired annular vortices appearing as "double doughnuts" (or tori) stacked axially one on top of the other around the rotor. The flow of the fluid in these vortices is, firstly, around the annulus or around the rotor, and, secondly, around the periphery of the "doughnut" between the rotor and stator as in a smoke ring (see Figure VI) and in such directions that any two sections of adjacent "doughnuts" rotate as though they were geared together. The width of the vortex in most cases corresponds to the distance between the rotor and stator so that the height of the "double doughnut" is at least about twice this distance.

Upon introducing another fluid substance into the continuous fluid in the contact zone, which substance forms a separate phase with the fluid in the zone, dispersion of the substance occurs. The dispersed particles are, more or less, carried in the paths of flow of the continuous phase in and around these vortices. The degree of dispersion and the degree to which the dispersed phase is carried in these vortices, as well as the rate of transfer from one phase to the other, depend upon (1) the magnitude of the shearing force in the continuous phase and (2) the relative properties of the continuous phase and dispersed phase. These factors are:

1. The relative angular velocity of rotation of adjacent contact surfaces.
 2. The radii of the contact surfaces.
 3. The rate of flow of the phases (continuous and dispersed) through the contact zone.
 4. The settling force either natural, that is by gravity, or induced, such as by electrostatic or magnetic fields.
 5. The roughness of the contactor surface.
 6. The temperature.
 7. The pressure.
 8. The dynamic stability of the apparatus.
 9. The viscosity of the phases.
 10. The density of the phases and the density differential between them.
 11. The interfacial tension between the phases.
 12. The adhesion tensions between the phases and cylindrical surfaces.
 13. The film coefficients of mass transfer between the two phases.
 14. The distribution of the distributed component between the phases.
 15. The pH of the system.
- The independent factors which affect the rate of transfer for two liquid phases in countercurrent flow for a given column and a given liquid system are: the speed of rotation, the rate of flow of each phase into the zone, the phase which is dispersed, the temperature, and the pressure.

The relative size of the particles of the dispersed phase to the size of the vortex is another factor in determining the degree to which the particles are carried by the vortex. Thus in contactors having the same rotor diameter to stator diameter ratio, particles of a given size are more easily carried in vortices in larger contactors than they would be in smaller contactors.

When the substance to be dispersed is a liquid or a gas, the shearing forces in the continuous phase cause it to be

broken up into particles of a limited size distribution, which size depends also upon the relative properties of the two phases. This means that the particles of the dispersed phase are sufficiently uniform in size and shape so that about 90% of the dispersed phase is in particles having sedimentation velocities which lie within the range of from about $\frac{1}{2}$ to 4 times the average sedimentation velocity (the average sedimentation velocity,

$$V = \frac{F_d}{H} + \frac{F_c}{1-H}$$

wherein H is the holdup or volume fraction of the dispersed phase in the contact zone, and F_c and F_d are the feed rates of the continuous and dispersed phase in volume per unit area per unit time, respectively). The particles of the dispersed phase remain at substantially the same size throughout their passage through the contact zone.

The formation of particles of uniform size has the advantage of preventing emulsification and/or foaming often caused by the formation of at least some exceedingly small sized particles in other types of contacting apparatus. Accordingly, the process of this invention is particularly applicable in contacting fluid systems which, in other apparatus, would give rise to emulsification difficulties.

As the relative difference in angular velocity between the two surfaces of the contact zone is increased, more and more particles of the dispersed phase are carried by the vortices. Different amounts of particles carried by the vortices of the continuous phase form different visual patterns which are a visual indication of the extent of contact between the dispersed phase and the continuous phase. Therefore, the pattern can be used as a way of estimating the extent of contact between the two phases.

There is a definite range of relative angular velocities within which efficient contacting is obtained and within which definite patterns are observed. This range of efficient contacting is the range of "orderly distribution" and particularly the region in this range of the "herringbone pattern." The regions and corresponding patterns, which are obtained as the relative angular velocity between the surfaces of the contact zone increases from zero up to and beyond the range of efficient contacting, may be described as follows:

Region	Apparent Size of the Particles of the Dispersed Phase	Apparent Distribution of the Dispersed Particles in the Continuous Phase	Apparent Flow of the Particles through the Annular Contact Zone	Apparent Pattern of the Particles in the Annular Contact Zone	See Figures	Efficiency of Contact between the Dispersed and Continuous Phases
A	Streamers and large particles	Irregular	Helical	Irregular	VIIa, b, c	Poor
B	Smaller particles and of more uniform size	Orderly distribution	From slightly pitched to horizontal	Striated	VIIIa VIIIb	Fair
C	Small uniformly sized particles	Orderly distribution	Horizontal around the rotor; particles concentrated in the vortices	Alternate regular periodic narrow bands of clear continuous phase and dispersed particles and wide bands where many particles are dispersed in the continuous phase, an incipient "herringbone" pattern	IXa IXb	Good
D	Small uniformly sized particles	Orderly distribution	Around the rotor in the vortices and around the sections of the vortices	Beautiful and clear "herringbones"	Xa Xb	Excellent
E	Small uniformly sized particles	Orderly distribution	Around the rotor in the vortices and around the sections of the vortices	Acute "herringbones" and may even appear banded as in Region C above	XIa XIb	Excellent
F	Small particles	Random turbulence	None, random	No orderly pattern		No controlled flow through the contact zone

Figures VIIa to XIb, consecutively, represent the different types of patterns obtained in an apparatus of the type disclosed in Figure V as the speed of the rotor is increased from very slow rotation up to the speed at which random turbulence occurs. Although Figures VIIa and VIIb show patterns observed at substantially the same rotor speed, the difference in appearance is due to the fact that, at this speed, changes in surface tensions have a large effect on the appearance of the pattern. The fluids represented in Figure VIIa have a higher interfacial tension than those represented in Figure VIIb. As the speed is increased still within the Region A, both the systems shown in Figures VIIa and VIIb will appear, more or less, like that shown in Figure VIIc.

Figure XII depicts the appearance of the dispersed phase in the continuous phase if an instantaneous photograph were taken of the contactor when operating in Regions C, D or E of the "doughnut" herringbone pattern.

With certain rotary contactors and certain fluid systems the "doughnut" type vortices are replaced by a double helix type pattern appearing like a rotating "barber pole" (see Figures XIIIa and XIIIb). In the double helix the particles move around the rotor and around the periphery of a section of the helix between the rotor and the stator (see Figure XIV) in such directions that two adjacent helix sections rotate as though they were geared together. The double helix may first appear in any one of the Regions C, D or E of "orderly distribution."

In Region F, random turbulence results in failure of the dispersed phase to travel independently in the direction of its gravitational force. In a counter-current system, this causes the column to "flood," that is, both phases are simultaneously withdrawn through either or both outlet lines 10 and 11 of Figure V. At the point of random turbulence the desired types of contact patterns disappear, and the advantages such as orderly flow, orderly distribution, minimum emulsifying tendencies, and controlled residence times are no longer obtained.

The point at which flooding occurs may be determined as follows:

Plot on the same graph (not shown) the two curves defined by (1) the ratio of the volume of dispersed phase to the volume of continuous phase in the column against the peripheral speed of the rotor, and (2) the ratio of the linear velocity of the dispersed phase to the continuous phase through the column against the

peripheral speed of the rotor. The point of intersection of curves (1) and (2) defines the point of flooding. The phenomenon of flooding may be represented by the following equation: 70

$$H = \frac{1}{\left(\frac{F_c}{F_d}\right)^{\frac{1}{2}} + 1}$$

wherein H is the holdup in the column when flooding occurs, and F_c and F_d are respectively the feed rates of the continuous and dispersed phases, expressed in volume per unit area per unit of time. 75

Another criterion for efficient contact in the rotary contactor of this invention may be expressed by a Reynolds number for rotational flow of the fluid mixture in the annular zone. This Reynolds number for rotational flow is represented by the formula: 80

$$R_o = \frac{(R_2 - R_1) V \rho}{\mu}$$

R_2 = radius of the outer surface of revolution 85

R_1 = radius of the inner surface of revolution

$(R_2 - R_1)$ = the clearance

$V = R_1 \omega_1$ when $\omega_2 = 0$; $V = R_2 \omega_2$ when $\omega_1 = 0$; and 90

$V = \frac{R_2 + R_1}{2} (\omega_2 - \omega_1)$ when both sur-

faces are rotating, wherein ω_2 is the angular velocity of the outer surface of revolution; ω_1 is the angular velocity of the inner surface of revolution, and ω_2 is not equal to ω_1 95

ρ = density of the fluid mixture in the annular contact zone

μ = viscosity of the fluid mixture in the annular contact zone. 100

When only the inner surface of revolution of the annular zone is rotated the product of the peripheral velocity times the clearance between the two surfaces at any section through the zone divided by 105

the kinematic viscosity $\frac{(\mu)}{(\rho)}$ of the fluid

mixture in the annular zone should be greater than about 2500 and preferably greater than about 3000; these values represent dimensionless numbers. 110

When only the outer surface of revolution is rotated a preferred condition of operation is when the product of the peripheral velocity times the clearance between the two surfaces of revolution at any section through the zone divided by 115

the kinematic viscosity of the fluid mixture in the zone is greater than about 2000, a dimensionless number. The critical value under these conditions is less than in the preceding instance because stable eddies are maintained over a wider range in this type of apparatus.

In an apparatus of the type shown in Figure V, when contacting aqueous systems wherein the kinematic viscosity of the fluid mixture in the column is not substantially greater than that of water, effective contact is obtained when the peripheral velocity of the inner cylinder is greater than about 2.5 or 3 feet per second.

In a contactor apparatus where both surfaces of revolution are rotated in the same direction the ratio of the angular velocity of the inner surface of revolution to the outer surface of revolution should be greater than the square of the ratio of the radius of said outer surface to the radius of said inner surface, that is, at any section through the contact

$$\frac{\omega_1}{\omega_2} > \left(\frac{R_2}{R_1} \right)^2$$

The relationship between the efficiency and the relative peripheral velocity of a rotor in an apparatus similar to that shown in Figure V for a liquid-liquid extraction system is illustrated by the curves shown in Figure XV. The several curves of the family shown in Figure XV represent different throughputs for a given column when a given one of the two phases is dispersed; the total throughputs for each of the curves 50, 51, 52 and 53 are .0231, .0385, .0665, and .135 gals./sq. in./min., respectively, thus the greater the throughput the greater the efficiency for the same peripheral velocity.

Dotted curve 54 is based on the same throughput as curve 52 but with the phases reversed. This curve 54, in comparison with curve 52, shows that either phase may be the dispersed phase and still give efficient extraction.

The curves terminate at the point where any further increase in peripheral velocity will cause flooding. All the curves go through a maximum provided they are not previously terminated by flooding. This maximum efficiency occurs at about the same peripheral speed for each different throughput for any given fluid system and in a given column (see Figure XV). The efficiency at the maximum of curve 50 can be obtained also on each of the curves 51, 52, and 53 at correspondingly higher throughputs and

lower peripheral velocities. These curves indicate the range of throughputs is very great, while at the same time reasonably high efficiency levels can be maintained. In other words, the operating range of the rotary contactor is very broad which is an important advantage in industrial processes. Thus, when driving the rotor with a variable speed motor many different effects may be easily obtained, and the efficiency of the contacting may be controlled at will within wide limits.

Changing the relative angular velocity of the surfaces in a given column is analogous to changing the type or size of the packing material, such as Raschig rings, and saddles in a packed tower; or the size of the orifice jets in a spray tower; or the hole size in a perforated plate tower. The ease of changing the speed is in marked contrast to the difficulty in changing the packing, jets or hole size in other columns.

Another important advantage of the rotary contactor of this invention is the ease of regulating residence time of the dispersed phase in the contact zone by controlling the relative peripheral velocity between the surfaces of the zone. This is particularly helpful in effecting the orderly procedure of all the dispersed particles through the zone.

In Figure XVI two curves were drawn from data obtained on a given aqueous liquid-liquid countercurrent extraction system wherein the two liquid phases had a minimum difference in density of about .2 gm./cc., a minimum interfacial tension of about 10 dynes/cm., and the viscosity of the continuous non-aqueous phase was about 1 centipoise. This system was contacted at different rotor speeds both in a large and in a small rotary contactor at several different total throughputs to obtain a series of curves having maxima like those shown in Figure XV. The efficiency of the contactor was obtained for each maxima at each throughput and these data for each contactor were plotted to obtain curves 60 and 61 in Figure XVI curve 60 being for the larger rotary contactor. According to the scale of total throughput in gallons per square inch of annular area per minute, good efficiency in such a system is obtained at a total throughput above about .025 and preferably above .05 gallons per square inch per minute. Between about .05 and .1 gallons per square inch per minute the optimum efficiency changes less than 20% for the corresponding 100% change in total throughput, and therefore the region above .05 gallons per square inch per minute is a preferred operating region

both because of the high efficiency and because of the wide permissible range in throughputs.

- In considering the effect of variations of size of the rotary contactor on its efficiency in comparison with similar variations of other contacting devices such as a packed or sieve plate tower, the following may be said: It is usually assumed by those skilled in the art that the stage efficiency for a contact tower such as a packed tower varies approximately inversely proportionally to the square root of the cross-sectional contact area. In Figure XVII the efficiency times the square root of the cross-sectional area is plotted against the effective cross-sectional area of different sized rotary contactors and packed towers for the same fluid system. If the efficiency were approximately proportional to the square root of the cross-sectional areas of both rotary contactors and the packed columns both lines 81 and 82 would be horizontal and parallel. But line 82 slopes upwardly as the size of the cross-sectional area of the contactor is increased, showing that with increasing size the rotary contactor is progressively more efficient than a packed tower. Also, the initial efficiency of a rotary contactor is materially greater than that of a packed tower of the same size, as illustrated by the fact that line 82 is materially above line 81. Rotary contactors are definitely superior to other types of contactors when they have effective annular cross-sectional areas of more than about 1.5 square inches. The lower the interfacial tension between the two phases, the easier it is to disperse one phase in the other. Therefore, surface-active agents may be added to either or both fluid phases to reduce the interfacial tension in the contact zone. Within limits, surface-active agents may also be used in this manner to prevent the formation of emulsions in the apparatus. The proper choice of a surface-active agent, either cation-active or anion-active, or both, depends upon the nature of the fluids being contacted. Some suitable surface-active agents are organic compounds containing one or more of the following radicals: metal or acid sulfate, sulfonate, phosphate, phosphonate, or carboxylate; hydroxyl or sulfhydryl, carbonyl, formyl, ether or ester; primary, secondary or tertiary amine, or quaternary ammonium and other onium base radicals; cyano, thiocyno or nitro; halide. For example, it is often desirable to use such agents in the extraction of penicillin from aqueous solutions with solvents to prevent emulsification of the

impurities in the penicillin solution.

Some illustrative uses for this invention are:

I. GAS-LIQUID AND LIQUID-GAS SYSTEMS.

General gas purification processes, gas stripping, including refluxing in distillation and extractive distillation, gas absorption, gas humidification and dehumidification.

II. GAS-SOLID SYSTEMS (wherein the solid is a powdered fluidized solid).

Adsorption operations such as the separation of methane and other light gases from ethane and ethylene in natural gas by adsorption on charcoal; adsorption reactions employing silica gel and other powdered adsorbent materials; fractionation of powder by elutriation or gas flotation. Chemi-adsorption operation such as the adsorption of olefins, diolefins or acetylenes by cuprous salts.

III. LIQUID-LIQUID SYSTEMS.

Solvent extraction processes for the separation of liquid mixtures containing organic and/or inorganic components of different degrees of polarity and/or saturation or molecular configuration such as the separation of olefins, diolefins, acetylenes, naphthenes, aromatics, alkyl phenols, thiophenols, and nitrogen bases from petroleum or coal tar oil fractions as well as their synthetic mixtures with one or more selective solvents such as sulfur dioxide, furfural, phenols, sulfolanes, copper-containing solutions, aldehydes, ketones, ethers, esters, amines, nitrites, and nitrohydrocarbons; the refining and purification of fatty acids, fatty oils, animal and vegetable oils and essential oils such as tung oil, linseed oil, soy bean oil, lemon oil, resin oil, tall oil, and cotton seed oil, with suitable solvents such as ethanolamine, furfural, and sulfolane; purification of dialkylamine such as diisopropylamine by its separation from the corresponding monoalkyl amine and other contaminants with a hydrocarbon solvent; the extraction, concentration and purification of antibiotics such as penicillin with selective solvents; the separation of polymers with solvents; the separation of epoxide and di-chlorohydrins from water with diisopropyl ether or other related solvent; and many other solvent extraction processes, particularly those wherein a large number of extraction stages is required because of small differences in solubilities of the two substances to be separated in the solvent employed for their separation. Neutralization reactions including liquid caustic alkali treating processes

such as the treatment of hydrocarbon oils containing acidic compounds such as hydrogen sulfide, mercaptans including thiophenols, alkyl phenols, naphthenic acids, sulfonic acids, by contact with aqueous caustic alkali solutions including solutizer solutions and the like; the separation of alkyl phenols from thiophenols and the separation of mixtures of alkyl phenols or of thiophenols into their components; the separation and purification of mixtures of sulfonic or naphthenic acids into their components. Acid treating processes such as the treatment of mineral oils with sulfuric acid to absorb gum forming components and other impurities are also included.

IV. LIQUID-SOLID SYSTEMS.

Leaching reactions such as the activation of clays with acid, the washing of precipitates, the deoiling of wax with solvents, the regeneration of adsorbents, the deoiling of adsorbents, the extraction of oil from shales, the extraction of oils from asphalts, the extraction of fatty oils from their meals or flakes, including cotton seeds, soy bean flakes, with organic solvents. The separation of proteins, including antigens, allergenic proteins, and other blood components, of hormones, vitamins, and antibiotics, from solutions containing them by adsorbing them on a solid adsorbent material. The separation of compounds of different degrees of polarity or saturation such as phenols from hydrocarbons, aromatics from non-aromatics, by absorption on a solid absorbing agent such as silica gel. Also included herein are processes involving ion exchange substances including ion exchange resins, and Permutite type substances.

V. LIQUID-GAS-SOLID SYSTEMS.

Mineral separation processes such as froth flotation processes.

VI. LIQUID-LIQUID-SOLID SYSTEMS.

The separation of proteins, including antigens, antibodies, allergenic proteins,

and other blood components, of hormones, vitamins and antibiotics from mixtures containing them by dissolving the mixture in a continuous phase and dispersing therein both adsorbing particles and an immiscible liquid phase whereby the adsorbing particles would be carried at the interface between the two liquid phases.

VII. LIQUID-LIQUID-LIQUID SYSTEMS.

Transfer of a solute from one solvent to a second solvent by contacting them countercurrently in a dispersing medium, such as extracting phenols from a high boiling hydrocarbon solvent into a low boiling denser solvent by contacting them countercurrently in an aqueous alkaline dispersing solution.

The following examples illustrate the effectiveness of several contacting processes in the apparatus by the method of this invention.

EXAMPLE I.

Gasolines having an ASTM 5% boiling point at 135° F. and a 90% boiling point at 338° F. and containing different amounts of tertiary butyl mercaptan were countercurrently contacted with a 3N aqueous solution of potassium hydroxide in two different sized glass contactor apparatus similar to that shown in Figure V. The apparatus of Runs 1 and 2 had an effective column height of 25 inches, a stator diameter of 1.312 inches, and a rotor diameter of 1 inch, while the apparatus of Runs 3 and 4 had a column height of 25 inches, a stator diameter of 2.185 inches and a rotor diameter of 1.476 inches. The runs were made with the following (see Table A below) different throughputs, different rotor speeds, and following degrees of extraction were obtained from which the efficiency in theoretical stages per foot of length was calculated according to the method described in the article by Hunter and Nash, *supra*. The caustic phase was the continuous phase and the gasoline phase formed the dispersed phase.

TABLE A.

Run No.	Throughput of KOH	Gals./in. ² / min. of Gasoline	% tert.-butyl mercaptan in feed as %	% tert.-butyl mercaptan in raffinate	Peripheral Velocity ft./sec.	Partition Coefficient of KOH to Gasoline	Temp. (° C.) of Contact	Efficiency in stages per ft.
1	.0040	.0195	.046	.0216	2.14	6.44	23.3	.422
2	.0039	.0096	.0470	.000432	3.30	6.53	23.0	5.61
3	.0011	.0054	.0479	.00817	4.65	6.36	23.8	1.67
4	.0018	.0088	.0500	.00169	4.65	6.53	23.0	3.45

Runs 1 and 2 show that with otherwise constant conditions an increase in peripheral speed to or above the neighborhood of 2.5—3 ft./sec. leads to a disproportionate increase in efficiency, while Runs 3 and 4 illustrate that where the peripheral speed is constant and the total throughput is increased by about 50% the efficiency is increased by slightly more than 100%. 5 10

A definite "herringbone" pattern was observed in the contactor during each of the above runs, except that of Run 1 which was operated at a peripheral speed below that required to obtain a pattern. 15

EXAMPLE II.

A narrow boiling fraction of petroleum alkyl phenols had an initial boiling point of 195.5° C., a final boiling point of 206.5° C., a 5% distillation temperature of 197° C., a 50% distillation temperature of 199° C., and a 95% distillation temperature of 205° C. An acetylation test showed that the mixture corresponds to about 57% xlenols and ethyl phenols (comprising substantial amounts of o-ethyl phenol, 2,6-, 2,4-, and 2,5-xlenols) and 43% of cresols (comprising substantial amounts of all three cresols). This mixture was dissolved in a sufficient amount of equal volumes of iso-octane and benzene to give a solution containing 200 grams of alkyl phenols per liter of solution. The resulting solution was extracted countercurrently with aqueous sodium hydroxide solution containing 2 moles per liter of NaOH in a rotary contactor similar to that shown in Figure V and having a rotor 25 inches long, a rotor diameter of 1.476 inches and a stator diameter of 2.185 inches. The solution of alkyl phenols was introduced at the rate of .01125 gals./in.²/minute and the caustic soda solution at the rate of .0053 gal./in.²/minute, the latter being the continuous phase. The rotor speed varied from 3.22 to 3.65 ft./sec. Well developed "herringbone" patterns were present under these operating conditions. 20 25 30 35 40 45

The xlenol and ethyl phenol content of the raffinate was 96%, and of the extract was 7.9%. 50

EXAMPLE III.

Several aqueous penicillin solutions were extracted with different solvents in a rotary contactor of the type shown in Figure V having a stator diameter of 2.0 inches, a rotor diameter of 1.3 inches and a length of 41 inches. The column was operated at between about 0° C. and 3° C. to prevent decomposition of the penicillin in the solution. The following data, in which the abbreviation MIBK is used to denote methylisobutylketone show the recovery and purity of penicillin obtained from different solutions under the following conditions: 55 60 65

TABLE B.

Run No.	Purity of Penicillin in aqueous feed solution Oxford Units/mg.* of solids	Solvent	Continuous Phase	Feed Ratio of Continuous: Dispersed Phase	Peripheral Velocity in ft./sec.	% Recovery of Penicillin in Extract	Purity of Penicillin in Oxford Units/mg.	
							Raffinate	Extract
1	175	MIBK	MIBK	1:1	5.68	97.5%	35	810
2	71	MIBK	Aqueous feed	1:1	3.69	92.1%	71	1043
3		50-50% CHCl ₃ -MIBK	CHCl ₃ -MIBK	0.67:1	5.68	99.5%		
4		CHCl ₃	CHCl ₃	0.67:1	6.2	95%		
5		CHCl ₃	Aqueous feed	0.67:1	3.12	96%		

*See article by Florey and Jennings in British Journal of Experimental Pathology, Vol. 23, p. 120, of June 1942.

A definite "herringbone" pattern was observed in the contactor during each of the above runs. Runs 4 and 5 in the above Table B show that efficient extraction can be obtained with two liquids of relatively great density difference (chloroform = 1.5, water = 1.0) when either phase is continuous.

EXAMPLE IV.

Two methyl isobutyl ketone solutions of acetic acid were contacted with water

in a contactor with a glass stator similar to that shown in Figure V at about room temperature at different rotor speeds and at different water-to-feed ratios. The percent of the acetic acid extracted by the water and the efficiency of the contactor in the number of theoretical stages per foot were determined for each run. Data on the two representative runs are shown in Table C below. The efficiencies were calculated according to the article mentioned in Example I above.

TABLE C.

Run No.	Dimension of Column		Concentration of acetic acid in Height feed moles/liter	% Acetic Acid in Raffinate	Extract moles/liter	Total Throughput gal./in. ² /min.	Feed Ratio Continuous: Dispersed	Peripheral vel. in ft./sec.	Efficiency stages/ft.	
	Stator diam.	Rotor diam.								
1	8.5	6.25	20	.526	0.091	.576	.065	1.32:1	10.2	1.41
2	8.5	6.25	20	0.466	0.084	0.518	.135	1.36:1	8.16	1.39

These examples indicate the flexibility of a rotary contactor and show that equal efficiency is obtained by change in peripheral velocity with a converse change in throughput.

A definite "herringbone" pattern was observed in the contact zone during each of the above runs in Table C.

EXAMPLE V.

Two different crude lubricating oil bulk distillates having viscosity indices below about 50 were contacted with furfural in a glass apparatus similar to

that shown in Figure V having a stator diameter of 3.75 inches, a rotor diameter of 2.14 inches, and 20 inches between feed ports along the column. The rotor was operated for each run at 300 RPM. The furfural was heated and introduced at the top of the column and the lubricating oil was heated and introduced at the bottom of the column. The raffinate phase of extracted oil was removed from the top of the column, freed of furfural, and tested for viscosity index V.I. The following table shows the conditions and results of each run:

Run No.	Lubricating Oil		Throughput in		Column Temp. ° C.		Raffinate	
	Feed Viscosity	S.U. sec. at 210° F.	cc./min.	Lube Oil Furfural	Top	Bottom	V.I.	% Yield
5 1	200		640	700	88	79	55	50
2	60		650	1075	96	69	44	41

In each of the runs in the above table the lube oil was the dispersed phase.

A "herringbone" pattern was not only observed in these runs, but also in runs where the furfural was the dispersed phase.

EXAMPLE VI.

A gaseous mixture of 27.1 vol. % H_2S and remainder of nitrogen was contacted countercurrently with an aqueous solvent solution containing 15.1% by weight of diethanolamine at atmospheric pressure and room temperature in a glass rotary contactor having a rotor diameter of 0.55 inches, a stator diameter of 0.75 inches and effective length of liquid therein of

28.0 inches. The rotor was rotated at a speed of 1395 RPM, the solvent was fed in at the top at a rate of .074 gal./hr. 25 and the gaseous mixture was fed in at the bottom at a rate of .425 cu. ft./hr. All of the H_2S was separated from the N_2 .

Comparative runs were made with the 30 same gaseous mixture but with an aqueous solvent solution containing only 6 wt. % diethanolamine. The rotary contactor employed had a glass stator 0.87 inches in diameter and a metal rotor of 0.63 35 inches in diameter. The following conditions and results of the runs were observed:

40	Rotor Speed	Liq. Ht. in inches	Inlet gas rate ft ³ /hr.	Solvent Rate Gal./hr.	H_2S conc. in inlet gas % v.	% H_2S Removed
	1240	33.0	.398	.039	24.5	72.5
	0	32.0	.398	.036	29.7	56.5

A pattern similar to that shown in Figure VIIIa was obtained when the rotor was rotated both at 1240 and 1395 RPM.

EXAMPLE VII.

A cracked light hydrocarbon gas boiling below the butane-butylene fraction and containing 2.8 v. % acidics, 25.2% H_2 , and 10 v. % products soluble in 98% H_2SO_4 containing 3% Ag_2SO_4 was concurrently contacted at atmospheric pressure and room temperature with powdered carbon black in a rotary contactor having a stainless steel rotor 0.63 inches in diameter, a glass stator 0.82 inches in diameter and an effective contact length of 26 inches. The rotor was operated at 2200 RPM, and the gas was fed into the contact zone at a rate of 120 gms./hr. The effluent gas contained 34.1 v. % H_2 , 0.6 v. % acidics, and 0.0% solubles in said acid. A definite "herringbone" pattern of the carbon particles was observed.

Specification No. 15381/46 (Serial No. 613,229) claims a rotary contactor apparatus for contacting at least two fluid substances which form at least two phases 70 when in contact with each other, com-

prising a substantially uninterrupted annular contact zone formed between two elongated structures having surfaces of revolution, one inside the other and out of contact with each other, wherein one of said surfaces is rotatable relative to the other, and shielding means in at least one end of said zones, mounted to permit relative rotation between the shield and other of said surfaces, providing a settling space between the shield and said other surface. I make no claim herein to a method of contacting which involves the use of apparatus including shielding means to provide an internal settling space as claimed in the said Specification.

Specification No. 15381/46 (Serial No. 615,479) claims a method of conducting a catalytic conversion in a heterogeneous mixture of phases which comprises passing the immiscible phases, one of which contains the catalyst, simultaneously and in suitable proportions under reaction conditions of temperature and pressure through a reaction zone substantially bounded by two spaced "relatively

- smooth surfaces" as therein described, said reaction zone being of cross-section and length to accommodate the desired throughput at the desired velocity with
- 5 non-turbulent flow and maintaining a condition of eddy flow as therein defined in said reaction zone by maintaining said boundary surfaces in a relative motion which is parallel for any point of one
- 10 surface and at least one corresponding point of the other surface. I make no claim herein to a method of contacting two or more substances when the latter include a catalyst.
- 15 Having now particularly described and ascertained the nature of my said invention and in what manner the same is to be performed (as communicated to me from abroad), subject to the foregoing
- 20 disclaimers, I declare that what I claim is:—
1. A method of contacting two or more substances none of which is a catalyst, which form at least two phases in contact
- 25 with each other, at least one of which substances is a fluid, comprising contacting said substances by passing said phases simultaneously, and preferably in counter current, into an annular zone
- 30 formed between two surfaces of revolution, one inside the other and out of contact with each other, wherein one of said surfaces rotates about its axis at a different angular velocity from that of
- 35 the other surface sufficient to maintain one phase dispersed in the other (continuous) phase and to maintain an orderly distribution, as hereinbefore defined of the dispersed phase in the continuous
- 40 phase.
2. The process according to claim 1, wherein the substances when contacted form at least a gas phase and a liquid phase.
- 45 3. The process according to claim 1, wherein the substances when contacted form a gas phase and a fluidized solid phase.
- 50 4. The process according to claim 1, wherein the substances when contacted form at least a liquid phase and a powdered solid phase.
5. The process according to claim 1, wherein the substances when contacted
- 55 form at least two liquid phases.
6. The process according to claim 1, wherein penicillin is recovered from a solution containing it by contact with a solvent at least partially immiscible with
- 60 said solution.
7. The process according to any of claims 1 to 6, wherein the dispersed phase is caused to form a herringbone

pattern in the annular zone.

8. The process according to any of claims 1 to 7, wherein the annular zone is formed by two coaxial cylindrical surfaces one of which cylindrical surfaces rotates about its axis at a different angular velocity from that of the other cylindrical surface.

9. The process according to any of claims 1 to 8, wherein the inner surface rotates about its axis at a greater angular velocity than that of the outer surface.

10. The process according to any of claims 1 to 9, wherein the relative peripheral velocity between the two surfaces of revolution forming the annular zone is greater than 2.5 feet per second, but is insufficient to produce random turbulence in said zone.

11. The process according to any of claims 1 to 9, wherein the relative peripheral velocity between the two surfaces of revolution forming the annular zone is greater than 2.5 feet per second, but is insufficient to produce flooding in said zone.

12. The process according to any of claims 1 to 9, wherein the inner surface rotates about its axis at a speed relative to that of the outer surface sufficient to maintain the ratio of the angular velocity of the inner surface to that of the outer surface greater than the square of the ratio of the radius of the outer surface to the radius of the inner surface at any section through the zone.

13. The process according to any of claims 1 to 11, characterized by the fact that a dissimilar substance which changes the surface-active properties of the fluid mixture in the annular zone is added thereto.

14. Penicillin whenever obtained or recovered by a process according to claim 6.

15. The process according to claim 1, wherein the outer surface is stationary and the inner surface rotates about its axis at an angular velocity sufficient to maintain the product of its peripheral velocity times the annular clearance between said surfaces of revolution at any section through the zone divided by the kinematic viscosity of the fluid mixture in the zone greater than 2500.

Dated the 5th day of February, 1946.

ELKINGTON & FIFE,

Consulting Chemists and Chartered
Patent Agents,
Bank Chambers,

329, High Holborn, London, W.C.1.
Agents for the Applicant.

[This Drawing is a reproduction of the Original on a reduced scale.]



Fig. 1a

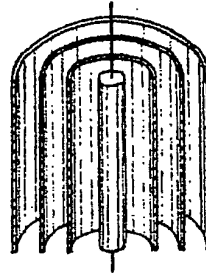


Fig. 1b

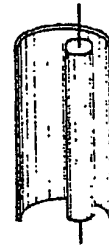


Fig. 1c



Fig. 1d

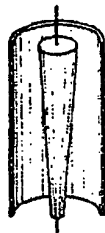


Fig. 2a



Fig. 2b



Fig. 2c

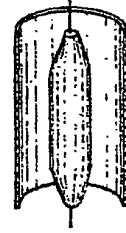


Fig. 2d



Fig. 3a

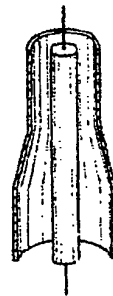


Fig. 3b



Fig. 3c

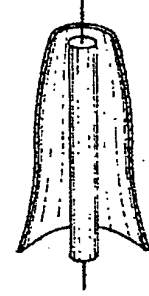


Fig. 3d

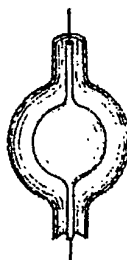


Fig. 4a



Fig. 4b

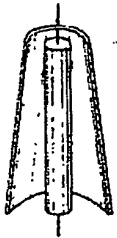


Fig. IIa

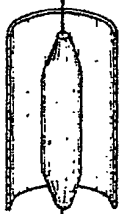


Fig. IIIa

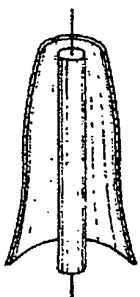


Fig. IVb

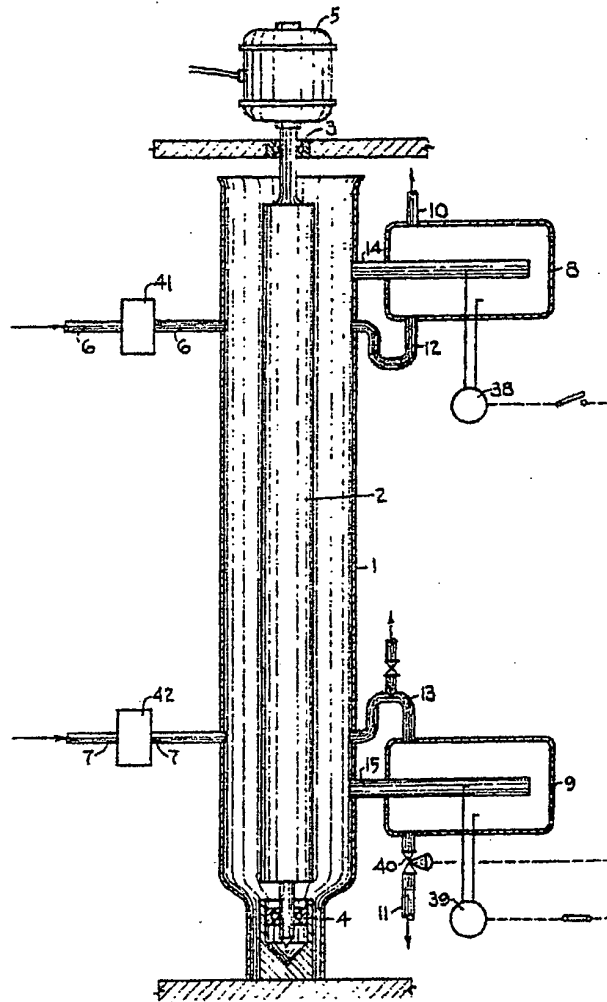
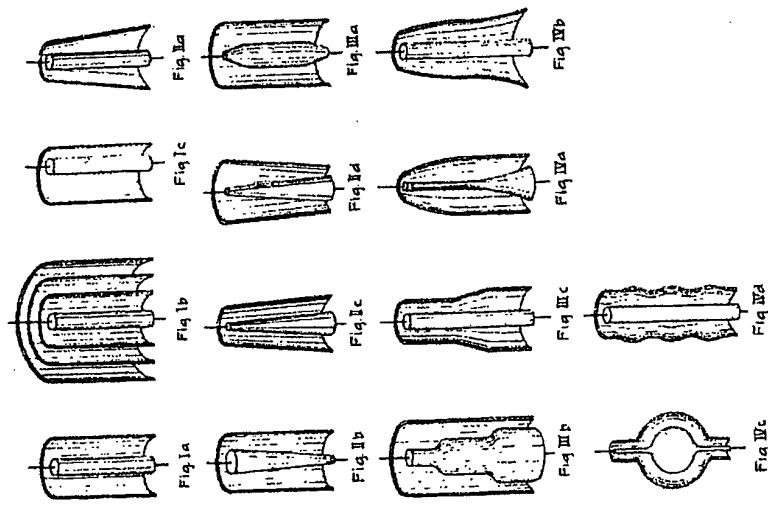


Fig. V



This Drawing is a reproduction of the Original on a reduced scale

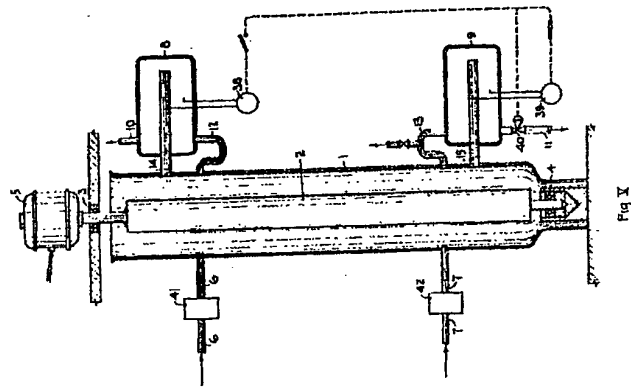


Fig. IX

[This Drawing is a reproduction of the Original on a reduced scale.]

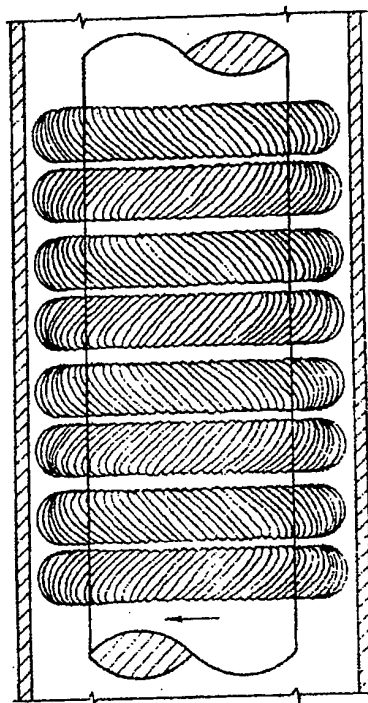


Fig. VI

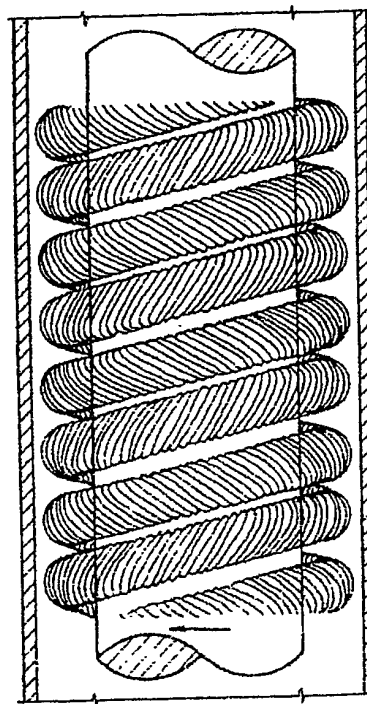
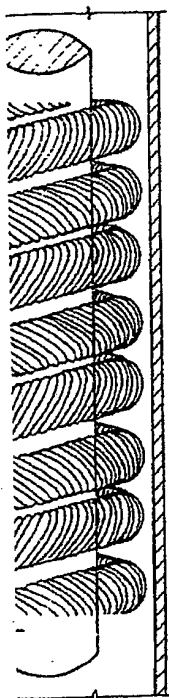


Fig. XIV



XIV

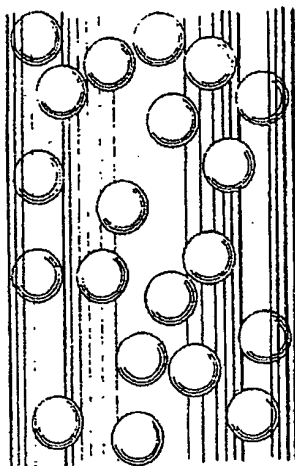


Fig. VIIa

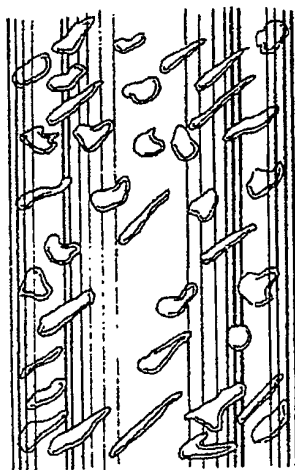


Fig. VIIb

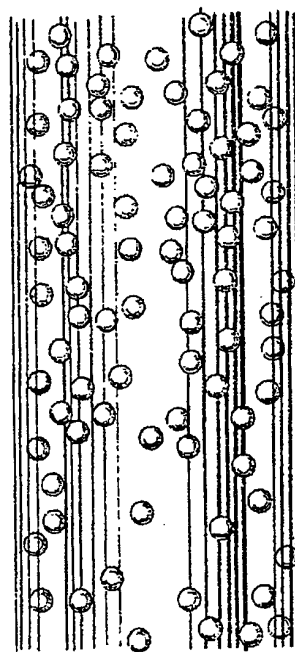


Fig. VIIc

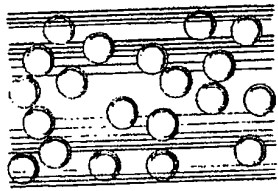


Fig. XIIa

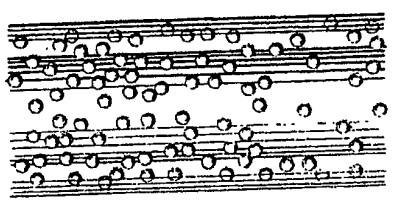


Fig. XIIc

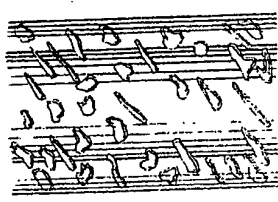


Fig. XIIb

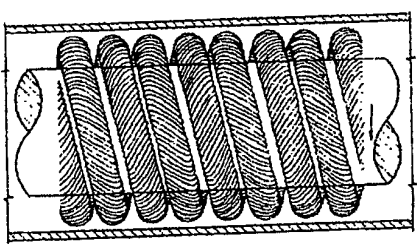


Fig. XIV

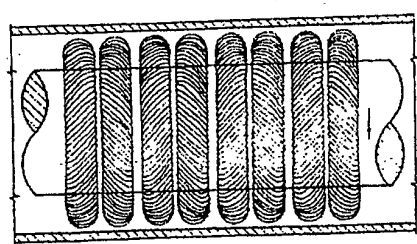


Fig. XVI

[This Drawing is a reproduction of the Original on a reduced scale.]

[This Drawing is a reproduction of the Original on a reduced scale.]

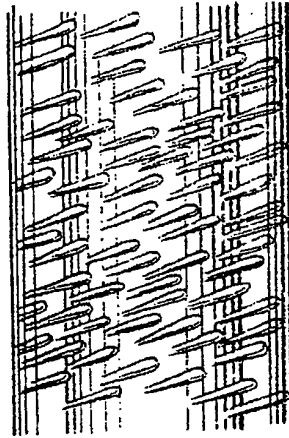


Fig. VIIa

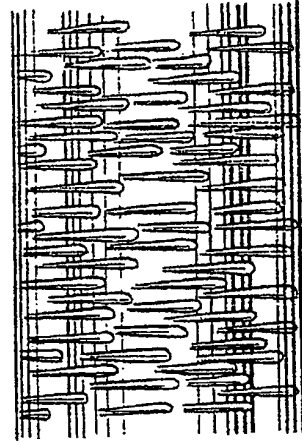


Fig. VIIIb

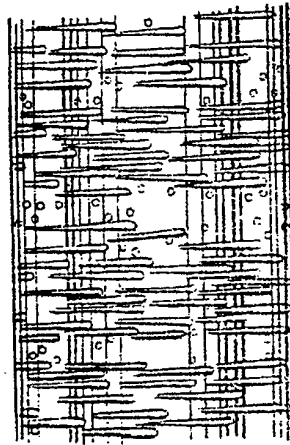


Fig. IXa

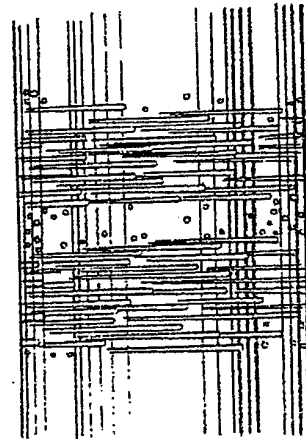


Fig. IXb

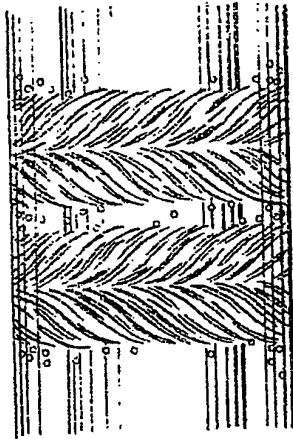
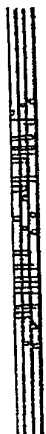
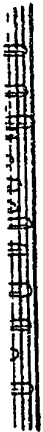


Fig. Xa

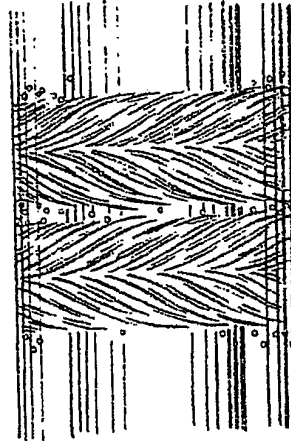


Fig. Xb

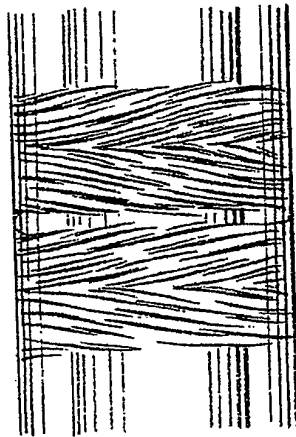


Fig. XIa

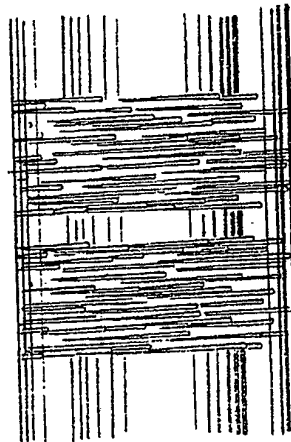


Fig. XIb

This Drawing is a reproduction of the Original on a reduced scale.

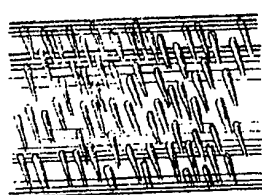


Fig. VIII a

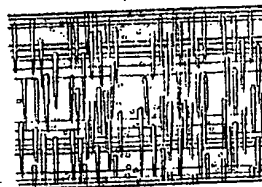


Fig IIa

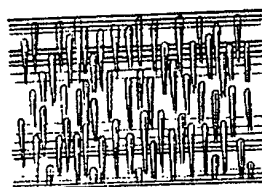


Fig. III b.

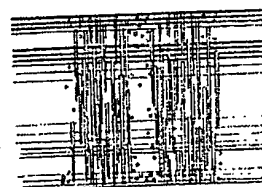


Fig IXb

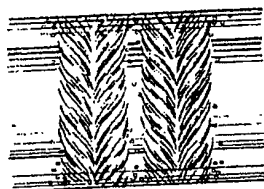
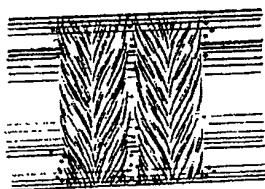


Fig. 2a



Fig 21a



98.5b

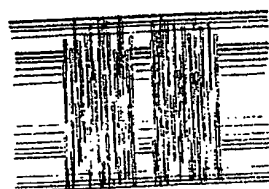


Fig. 21b

[This Drawing is a reproduction of the Original on a reduced scale.]

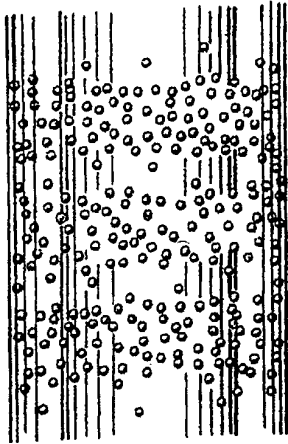


Fig. XII

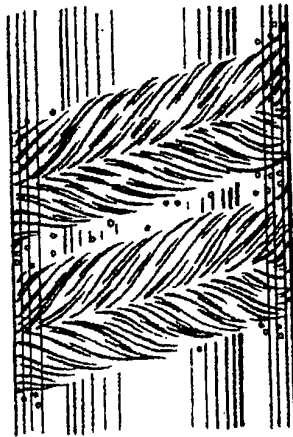
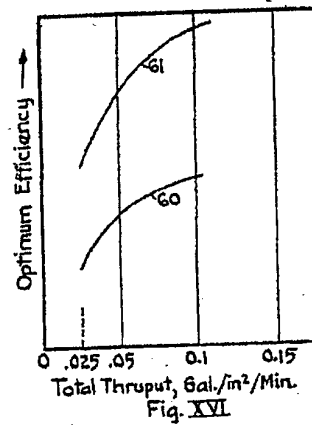
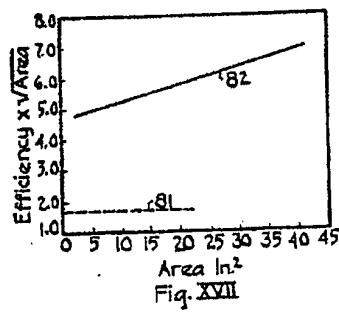
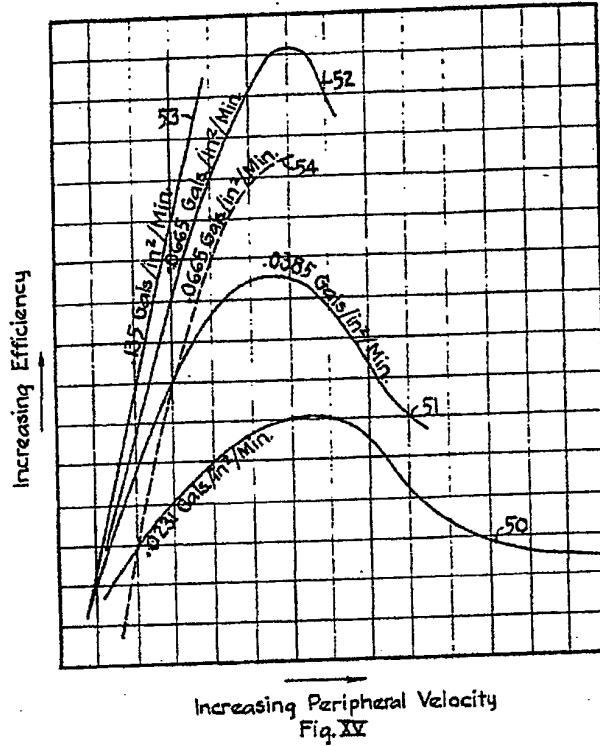


Fig. XIIIa



Fig. XIIIb



615425 COMPLETE SPECIFICATION

SHEET 7

SHEET 6

[This Drawing is a reproduction of the Original on a reduced scale]

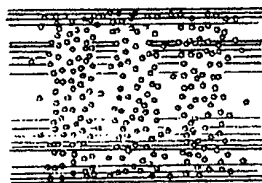


Fig. XII



Fig. XIIIb

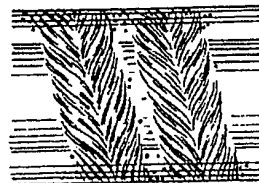


Fig. XIIIa

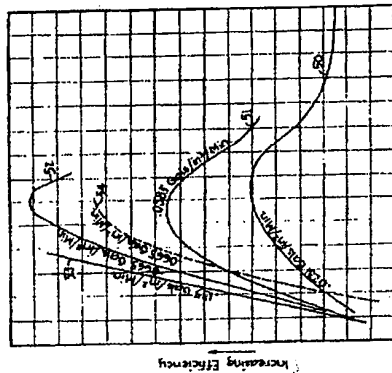


Fig. XI

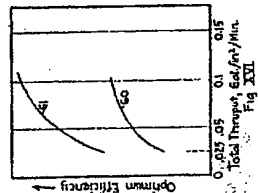


Fig. XXI

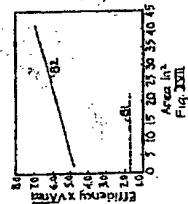


Fig. XII

615425A

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☐ **FADED TEXT OR DRAWING**
- ☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.